

Scanned 2/9/2005
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Japanese Patent Laid-Open No. 2001-52870

Laid-Open Date: February 23, 2001

Application No. 345071/1999

Application Date: December 3, 1999

Convention Priority No. 157176/1999

Convention Priority Date: June 3, 1999

Convention Priority Country: Japan (JP)

Applicant: TDK Corporation

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[Title of the Invention] ORGANIC EL ELEMENT

[Abstract]

[Object] It is an object of the present invention to provide an organic element which efficiently emits light of blue color, is applicable to the multi-color emission, including blue color, high in luminance and has long service life.

[Construction] The organic EL element of the present invention is provided with the blue-emitting layer containing the hole injecting/transferring compound in the hole transferring layer 4 and/or electron injecting/transferring compound in the electron transferring layer 7 as the host compound(s), these layers being adjacent to the light-emitting layer, or the blue-emitting layer combined with the cathode with a halide or oxide of an alkali metal as the constituent material.

Scanned 2/9/2005
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2/9/2005

[Claims]

[Claim 1] An organic EL element comprising a light-emitting layer, and hole and/or electron transferring layers adjacent to the light-emitting layer, wherein the light-emitting layer is composed of one or more layers including a blue-emitting layer which contains a hole injecting/transferring compound in the hole transferring layer and/or electron injecting/transferring compound in the electron transferring layer as the host compound(s).

[Claim 2] The organic EL element according to Claim 1, wherein said host compound emits blue light.

[Claim 3] The organic EL element according to Claim 2, wherein said host compound is selected from phenyl anthracene derivatives.

[Claim 4] The organic EL element according to the Claim 1, which contains a dopant by which it emits blue color.

[Claim 5] The organic EL element according to Claim 1, which has the hole and electron transferring layer, wherein the blue-emitting layer is a mixed layer of the hole injecting/transferring compound in the hole transferring layer and electron injecting/transferring compound in the electron transferring layer.

[Claim 6] The organic EL element according to Claim 5, wherein said blue-emitting layer is a mixed layer of a phenyl anthracene derivative and aromatic tertiary amine.

SECRET 2049/23885
3 9/8/2004

[Claim 7] The organic EL element according to Claim 5 or 6, wherein said hole injecting/transferring compound and electron injecting/transferring compound are contained each at a constant concentration in said mixed layer.

[Claim 8] The organic EL element according to Claim 5 or 6, wherein each of said hole injecting/transferring compound and electron injecting/transferring compound has a concentration distribution in the film thickness direction, said hole injecting/transferring compound having a higher concentration on the hole transferring layer side and said electron injecting/transferring compound having a higher concentration on the electron transferring layer side.

[Claim 9] The organic EL element according to one of Claims 5 to 8, wherein said mixed layer is doped with a dopant.

[Claim 10] The organic EL element according to one of Claims 5 to 9, wherein said mixed layer as a whole emits blue color.

[Claim 11] The organic EL element according to one of Claims 1 to 10, wherein the materials that constitute the cathode provided on said electron transferring layer side include at least one type of compound selected from the group consisting of a halide and oxide of an alkali metals.

[Claim 12] The organic EL element according to Claim 11, wherein the materials that constitute the cathode provided on said electron transferring layer side include at least one

type of compound selected from the group consisting of halides of Rb and Cs.

[Claim 13] An organic EL element comprising a cathode, one or more light-emitting layers including a blue-emitting layer, hole transferring and/or injecting layers, and anode, wherein the materials that constitute the cathode include at least one type of compound selected from the group consisting of a halide and oxide of an alkali metals.

[Claim 14] The organic EL element according to Claim 13, wherein said blue-emitting layer contains a phenyl anthracene derivative as the compound that emits blue color.

[Claim 15] The organic EL element according to Claim 13 or 14, wherein said hole transferring and/or injecting layers contain an aromatic tertiary amine.

[Claim 16] The organic EL element according to Claim 15, wherein said aromatic tertiary amine is selected from the group consisting of the compounds represented by the general formula (1) or (2):

[Formula 1]

(1)

wherein, R_1 , R_2 , R_3 and R_4 are each an aryl, alkyl, alkoxy, aryloxy or halogen group; r_1 , r_2 , r_3 and r_4 are each an integer of 0 to 5; when r_1 , r_2 , r_3 and r_4 are each an integer of 2 or more, adjacent R_1 's may be bonded to each other to form a ring, and

so may be R_2 's, R_3 's and R_4 's; R_5 and R_6 are each an alkyl, alkoxy, amino or halogen group, and r_5 and r_6 are each an integer of 0 to 4, and

[Formula 2]

(2)

wherein, ϕ is phenylene group; R_{01} , R_{02} , R_{03} and R_{04} are each an alkyl, aryl, diarylaminoaryl group, or

[Formula 3]

(3)

wherein, R_{011} , R_{012} , R_{013} , R_{014} , R_{015} , R_{016} and R_{017} are each an aryl group; at least one of R_{01} to R_{04} is diarylaminoaryl group, or one of (a-1) to (a-3); r_{01} , r_{02} , r_{03} and r_{04} are each an integer of 0 to 5; $r_{01}+r_{02}+r_{03}+r_{04}$ is an integer of 1 or more; when r_{01} , r_{02} , r_{03} and r_{04} are each an integer of 2 or more, adjacent R_{01} 's may be bonded to each other to form a ring, and so may be R_{02} 's, R_{03} 's and R_{04} 's.

[Claim 17] The organic EL element according to Claim 15 or 16 which has the hole injecting and transferring layers, the hole injecting layer on said anode side containing the compound represented by the formula (2) and hole transferring layer on said light-emitting layer side containing the compound represented by the formula (1).

[Claim 18] The organic EL element according to one of

Claims 1 to 17 which has at least one light-emitting layer, in addition to said blue-emitting layer, emitting light of wavelength different from that of the light emitted by said blue-emitting layer.

[Claim 19] The organic EL element according to Claim 18, wherein said at least one light-emitting layer emitting light of wavelength different from that of the light emitted by said blue-emitting layer is a mixed layer of the hole injecting or transferring compound and electron injecting or transferring compound.

[Claim 20] The organic EL element according to Claim 19, wherein said mixed layer is doped with a dopant.

[Claim 21] The organic EL element according to one of Claims 18 to 20 which has 2 light-emitting layers.

[Claim 22] The organic EL element according to one of Claims 18 to 20 which has 3 light-emitting layers.

[Claim 23] The organic EL element according to Claim 21 or 22 which emits white color.

[Claim 24] The organic EL element according to one of Claims 1 to 23 which is combined with a color filter to modulate emitted light.

[Claim 25] The organic EL element according to Claim 24 which includes a pair of electrodes facing each other and at least one of which is transparent, wherein the organic layer containing said light-emitting layer is placed between the

electrodes, and said color filter is provided on the side of the transparent electrode of the pair of electrodes.

[Claim 26] The organic EL element according to Claim 24 which includes a pair of XY matrix type electrodes at least one of which is transparent, each being composed of two or more electrodes, intersecting with each other and being positioned to face each other, wherein the organic layer containing said light-emitting layer is placed between the electrodes intersecting with each other, each intersection forms the picture element, and said color filter is provided on the transparent electrode side of the picture element.

[Claim 27] The organic EL element according to Claim 26, wherein a black matrix is provided around said picture element and in the vicinity of the position at which said color filter is provided.

[DETAILED DESCRIPTION OF THE INVENTION]

[0001]

[FIELD OF THE INVENTION]

This invention relates to an organic EL (electrical field luminescent) element.

[DESCRIPTION OF THE RELATED ART]

An organic EL element is a device to emit light, with a thin film containing a fluorescent organic compound placed between the cathode and anode, where the electrons and holes are injected into the thin film to be recombined with each other

to generate excitons, which emit light (fluorescence or phosphorescence) while being deactivated.

[0003]

An organic EL element can emit light of high luminance of around 100 to 100,000cd/m² from the surface at a low voltage of 10V or less. It can also emit light of widely varying colors from blue to red by selecting type of the fluorescent substance.

[0004]

An organic EL element has several disadvantages, e.g., short light-emitting life, and low storage durability and reliability, which result from the following causes.

(1) Physical changes of the organic compound

The interface becomes uneven, resulting from growth of the crystal domain or the like, which may cause the problems, e.g., deteriorated element capacity of injecting charges, short-circuit and insulation breakdown. Generation and growth of the grains, occurring especially when a low-molecular-weight compound having a molecular weight of 500 or less is used, notably deteriorates the film characteristics. The grains are also notably generated and grown, when the interfaces, e.g., that with ITO used for the anode, are roughened, to cause deteriorated light-emitting efficiency, current leakage, and eventually failure of the light-emitting capacity. It may also cause dark spots, which partially emit no light.

[0005]

(2) Oxidation and separation of the cathode

A metal of low work function, e.g., Na, K, Li, Mg, Ca or Al, is used for the cathode to facilitate injection of the electrons. These metals, however, react with moisture or oxygen in the air to cause separation of the cathode from the organic layer, preventing charges from being injected.

In particular, when the film of a high-molecular-weight compound is formed by spin coating, residual solvent, moisture and decomposed products left during the film-making step accelerate oxidation of the electrode, to cause separation of the electrode and generation of the dark spots.

[0006]

(3) Low light-emitting efficiency, and large exotherm

The organic compound is exposed to a high field strength, because electric current is passed through the compound, and invariably generates heat. The resultant heat may cause melting, crystallization and thermal decomposition of the organic compound, and hence deterioration and breakdown of the element.

[0007]

(4) Photochemical or electrochemical changes of the organic compound layer

The electric current passed through the organic compound may deteriorate the compound, causing defects, e.g., current and exciton trapping, and deterioration of the element, e.g., increased driving voltage and lowered luminance.

[0008]

The organic EL element can realize multi-color emission, as described above, and a laminate type white color emitting organic EL element is proposed as the one for multi-color emission (Yoshiharu Sato, Technical Reports of Shinshu University, OME-94-78 (1995-03)). The laminate is composed of the light-emitting layers of an oxazole complex of zinc for blue, tris(8-quinolinolate)aluminum for green and tris(8-quinolinolate) aluminum doped with a red fluorescent pigment (P-660, DCM1) for red.

[0009]

The inventors of the present invention have proposed the technique for multi-color emission with the aid of dopants (WO98/08360) in consideration of greatly limited material selection and freedom of controlling emitted light involved in the above element. More specifically, a mixed layer of tris(8-quinolinolate)aluminum and N,N,N',N'-tetrakis-(3-biphenyl-1-yl) benzidine is doped with rubrene or coumarin derivative. It can realize multi-color emission by changing the mixing ratio of the components and dopant species.

[0010]

However, the specifically disclosed colors emitted correspond to red to green, but not to blue.

[0011]

Therefore, stable production of blue color is in demand, but involves specific problems. It is necessary to select adequate materials, not only for the light-emitting ones but also for those combined therewith.

[0012]

[PROBLEMS THAT THE INVENTION IS TO SOLVE]

It is an object of the present invention to provide a highly reliable organic EL element capable of efficiently emitting blue color. It is another object of the present invention to provide an organic EL element which can cope with emission of multi-colors including blue, produce colors of high luminance, and is serviceable for extended periods. It is still another object of the present invention to provide an organic EL element which can be used for a multi-color emitting organic EL display with its excellent characteristics by being combined with a color filter.

[0013]

[MEANS FOR SOLVING THE PROBLEMS]

The above objects can be achieved by the inventions, described below.

- (1) An organic EL element comprising a light-emitting layer, and hole and/or electron transferring layers adjacent to the light-emitting layer, wherein the light-emitting layer is composed of one or more layers including a blue-emitting layer which contains a hole injecting/transferring compound in the

hole transferring layer and/or electron injecting/transferring compound in the electron transferring layer as the host compound(s).

(2) The organic EL element according to the invention (1), wherein the host compound emits blue color.

(3) The organic EL element according to the invention (2), wherein the host compound is selected from phenyl anthracene derivatives.

(4) The organic EL element according to the invention (1), which contains a dopant by which it emits blue color.

(5) The organic EL element according to the invention (1), which has a hole and electron transferring layer, wherein the blue-emitting layer is a mixed layer of the hole injecting/transferring compound in the hole transferring layer and electron injecting/transferring compound in the electron transferring layer.

(6) The organic EL element according to the invention (5), wherein the blue-emitting layer is a mixed layer of a phenyl anthracene derivative and aromatic tertiary amine.

(7) The organic EL element according to the invention (5) or (6), wherein the hole injecting/transferring compound and electron injecting/transferring compound are contained each at a constant concentration in a mixed layer.

(8) The organic EL element according to the invention (5) or (6), wherein each of the hole injecting/transferring compound

and electron injecting/transferring compound has a concentration distribution in the film thickness direction, the hole injecting/transferring compound having a higher concentration on the hole transferring layer side and the electron injecting/transferring compound having a higher concentration on the electron transferring layer side.

(9) The organic EL element according to one of the inventions (5) to (8), wherein the mixed layer is doped with a dopant.

(10) The organic EL element according to one of the inventions (5) to (9), wherein the mixed layer as a whole emits blue color.

(11) The organic EL element according to one of the inventions (1) to (10), wherein the materials that constitute the cathode provided on the electron transferring layer side include at least one type of compound selected from the group consisting of a halide and oxide of an alkali metals.

(12) The organic EL element according to the invention (11), wherein the materials that constitute the cathode provided on the electron transferring layer side include at least one type of compound selected from the group consisting of halides of Rb and Cs.

(13) An organic EL element comprising a cathode, one or more light-emitting layers including a blue-emitting layer, hole transferring and/or injecting layers, and anode, wherein the materials that constitute the cathode include at least one type of compound selected from the group consisting of a halide and

oxide of an alkali metals.

(14) The organic EL element according to the invention (13), wherein the blue-emitting layer contains a phenyl anthracene derivative as the compound that emits blue color.

(15) The organic EL element according to the invention (13) or 14, wherein the hole transferring and/or injecting layers contain an aromatic tertiary amine.

(16) The organic EL element according to the invention (15), wherein the aromatic tertiary amine is selected from the group consisting of the compounds represented by the general formula (1) or (2):

[0014]

[Formula 4]

(1)

[0015]

wherein, R_1 , R_2 , R_3 and R_4 are each an aryl, alkyl, alkoxy, aryloxy or halogen group; r_1 , r_2 , r_3 and r_4 are each an integer of 0 to 5; when r_1 , r_2 , r_3 and r_4 are each an integer of 2 or more, R_1 's may be bonded to each other to form a ring, and so may be R_2 's, R_3 's and R_4 's; R_5 and R_6 are each an alkyl, alkoxy, amino or halogen group, and r_5 and r_6 are each an integer of 0 to 4, and

[0016]

[Formula 5]

(2)

[0017]

wherein, ϕ is phenylene group; R_{01} , R_{02} , R_{03} and R_{04} are each an alkyl, aryl, diarylaminoaryl group, or

[0018]

[Formula 6]

(a-1) (a-2) (a-3)

wherein, R_{011} , R_{012} , R_{013} , R_{014} , R_{015} , R_{016} and R_{017} are each an aryl group;

[0019]

at least one of R_{01} to R_{04} is diarylaminoaryl group, or one of (a-1) to (a-3); r_{01} , r_{02} , r_{03} and r_{04} are each an integer of 0 to 5; $r_{01}+r_{02}+r_{03}+r_{04}$ is an integer of 1 or more; when r_{01} , r_{02} , r_{03} and r_{04} are each an integer of 2 or more, adjacent R_{01} 's may be bonded to each other to form a ring, and so may be R_{02} 's, R_{03} 's and R_{04} 's.

(17) The organic EL element according to the invention (15) or (16) which has the hole injecting and transferring layers, the hole injecting layer on the anode side containing the compound represented by the formula (2) and hole transferring layer on the light-emitting layer side containing the compound represented by the formula (1).

(18) The organic EL element according to one of the inventions

(1) to (17) which has at least one light-emitting layer, in addition to the blue-emitting layer, emitting light of wavelength different from that of the light emitted by the blue-emitting layer.

(19) The organic EL element according to the invention (18), wherein the at least one light-emitting layer emitting light of wavelength different from that of the light emitted by the blue-emitting layer is a mixed layer of the hole injecting or transferring compound and electron injecting or transferring compound.

(20) The organic EL element according to the invention (19), wherein the mixed layer is doped with a dopant.

(21) The organic EL element according to one of the inventions (18) to (20) which has 2 light-emitting layers.

(22) The organic EL element according to one of the inventions (18) to (20) which has 3 light-emitting layers.

(23) The organic EL element according to the invention (21) or (22) which emits white color.

(24) The organic EL element according to one of the inventions (1) to (23) which is combined with a color filter to modulate emitted light.

(25) The organic EL element according to the invention (24) which includes a pair of electrodes facing each other and at least one of which is transparent, wherein the organic layer containing the light-emitting layer is placed between the

electrodes, and the color filter is provided on the side of the transparent electrode of the pair of electrodes.

(26) The organic EL element according to the invention (24) which includes a pair of XY matrix type electrodes at least one of which is transparent, each being composed of two or more electrodes, intersecting with each other and being positioned to face each other, wherein the organic layer containing the light-emitting layer is placed between the electrodes intersecting with each other, each intersection forms the picture element, and the color filter is provided on the transparent electrode side of the picture element.

(27) The organic EL element according to the invention (26), wherein a black matrix is provided around the picture element and in the vicinity of the position at which the color filter is provided.

[0020]

[DESCRIPTION OF THE PREFERRED EMBODIMENTS]

The present invention is described in more detail. The organic EL element of the present invention includes a blue-emitting layer which contains, as the host compound(s), a hole injecting/transferring compound and/or electron injecting/transferring compound in the hole transferring layer and/or electron transferring layer adjacent to the light-emitting layer, or includes a blue-emitting layer and uses a compound selected from the group consisting of a chloride

and oxide of an alkali metal as the material for the cathode. It preferably has both these structures with the blue-emitting layer preferably being of a mixed layer of the hole and electron injecting/transferring compounds. The present invention is described in still more detail.

[0021]

<Blue-emitting layer>

The organic EL element of the present invention includes a blue-emitting layer, wherein a phenylene anthracene derivative is the preferable compound for emitting blue color. The phenylene anthracene derivatives useful for the present invention are described in Japanese Patent Laid-open No. 12600/1996. Of these, those represented by the formula (A) are more preferable:

A_1-L-A_2 (A)

[0022]

wherein, A_1 and A_2 are each mono(ortho-substituted phenyl)anthryl or di(ortho-substituted phenyl)anthryl group, which may be the same or different; and L is a single bond or divalent coupling group.

[0023]

The mono(ortho-substituted phenyl)phenyl anthryl or di(ortho-substituted phenyl)phenyl anthryl group represented by A_1 , A_2 in the formula (A) has an aryl, aromatic heterocyclic or arylolethenyl group at the 2- or 6-site of the phenyl group

(ortho-site with respect to the position at which it is bonded to the anthracene ring). It may have a substituent at a site other than the ortho-site. Examples of the substituent, when bonded, include alkyl, aryl, arylothenyl, alkoxy and amino group. They may be further substituted. These substituents are described later.

[0024]

The phenyl group is preferably bonded to the anthracene ring at the 9- or 10-site.

[0025]

L in the formula (A) is a single bond or divalent group, and the divalent bond is preferably an arylene group which may include an alkylene group or the like. These arylene groups are described later.

[0026]

Of the phenyl anthracene derivatives represented by the formula (A), those represented by the formula (A-1) or (A-2) are more preferable.

[0027]

[Formula 7]

(A-1)

[0028]

[Formula 8]

(A-2)

[0029]

In the formula (A-1), Ar₁ to Ar₄ are each hydrogen atom, or an aryl, aromatic heterocyclic or arylothenyl group, at least one of Ar₁ and Ar₂, and at least one of Ar₃ and Ar₄ are an aryl, aromatic heterocyclic or arylothenyl group; R₅₁ and R₅₂ are each an alkyl, aryl, arylothenyl, alkoxy or amino group, which may be the same or different; p₁ and p₂ are each an integer of 0 to 3, when p₁ or p₂ is an integer of 2 or more, R₅₁'s or R₅₂'s may be the same or different; R₅₃ is an alkyl or aryl group; p₃ is an integer of 0 to 3, when p₃ is an integer of 2 or more, R₅₃'s may be the same or different; and L₁ is a single bond or arylene group, which may include an alkylene group, -O-, -S- or -NR- (R is an alkyl or aryl group).

[0030]

In the formula (A-2), Ar₅ and Ar₆ are each hydrogen atom, or an aryl, aromatic heterocyclic or arylothenyl group, at least one of Ar₅ and Ar₆ is an aryl, aromatic heterocyclic or arylothenyl group; R₅₄'s are each an alkyl, aryl, arylothenyl, alkoxy or amino group, which may be the same or different; p₄ is an integer of 0 to 3, when p₄ is an integer of 2 or more, R₅₄'s may be the same or different; R₅₅ is an alkyl or aryl group; p₅ is an integer of 0 to 4, when p₅ is an integer of 2 or more, R₅₅'s may be the same or different; and L₂ is a single bond or arylene group, which may include an alkylene group, -O-, -S- or -NR- (R is

an alkyl or aryl group).

[0031]

The aryl groups represented by Ar_1 to Ar_4 or R_{51} to R_{53} preferably have 6 to 20 carbon atoms, and may have a substituent, e.g., phenyl or tolyl. More specifically, the substituent may be phenyl, o-, m- or p-tolyl, pyrenyl, naphthyl, anthryl, biphenyl, phenylanthryl, tolylanthryl, or the like.

[0032]

The preferable aromatic heterocyclic groups represented by Ar_1 to Ar_4 include furyl, benzofuryl, thienyl, bithienyl, benzothienyl, pyrrolyl, N-allylpyrrolyl, indolyl, pyridyl, bipyridyl, quinolyl, quinoxalyl, oxazole, benzoxazole, oxadiazole, thiazole, benzothiazole, thiadiazole and imidazole. Moreover, they may have a substituent, e.g., an aryl group of 42 or less carbon atoms, or alkyl, alkoxy, aryloxy, amino, cyano or nitro group of 12 or less carbon atoms. More specifically, the substituent may be phenyl, o-, m- or p-biphenyl, (1,2)naphthyl, methyl, ethyl, propyl, butyl, methoxy, ethoxy, phenoxy, or o-, m- or p-tolyl group.

[0033]

The preferable arylolethenyl groups represented by Ar_1 to Ar_4 or R_{51} or R_{52} include 2-phenylethenyl and 2,2-diphenylethenyl. They may have a substituent, e.g., an aryl, alkyl, alkoxy, aryloxy, amino, cyano or nitro group. More specifically, the substituent may be phenyl, o-, m- or p-biphenyl, (1,2)naphthyl,

methyl, ethyl, propyl, butyl, methoxy, ethoxy, phenoxy, or o-, m- or p-tolyl group.

[0034]

The alkyl groups represented by R_{51} to R_{53} may be of straight-chain or branched having 1 to 10 carbon atoms, preferably 1 to 4 carbon atoms, and may be substituted or not substituted. Still more preferably, they have 1 to 4 carbon atoms, and are not substituted. More specifically, they include methyl, ethyl, n- and i- propyl, and n-, i-, s- and t-butyl group.

[0035]

The alkoxy groups represented by R_{51} or R_{52} preferably has the alkyl group section of 1 to 6 carbon atoms. More specifically, they include methoxy and ethoxy group. The alkoxy group may be substituted.

[0036]

The amino group represented by R_{51} or R_{52} may be substituted or not substituted, the former being more preferable. The substituents useful for the present invention include an alkyl (e.g., methyl and ethyl) and aryl (e.g., phenyl) group, more specifically diethylamino, diphenylamino and di(m-tolyl)amino group.

[0037]

In the formula (A-1), p_1 and p_2 are each an integer of 0 to 3, particularly preferably 0 to 2. When p_1 and p_2 are each

an integer of 1 to 3, particularly 1 or 2, R_{51} and R_{52} are each preferably methyl or phenyl.

[0038]

In the formula (A-1), p_3 is each an integer of 0 to 3, particularly preferably 0 to 2. When p_3 is each an integer of 1 to 3, particularly 1 or 2, R_{53} are each preferably methyl or phenyl.

[0039]

In the formula (A-1), R_{51} to R_{53} may be the same or different. When there are two or more R_{51} 's, they may be the same or different, and so may be R_{52} 's and R_{53} 's.

[0040]

In the formula (A-1), L_1 is a single bond or arylene group. The arylene groups represented by L_1 are preferably not substituted, and specifically include not only the common arylene groups, e.g., phenylene, biphenylene and anthrylene, but also those with two or more arylene groups directly bonded to each other. L_1 is preferably a single bond, p-phenylene or 4,4'-biphenylene.

[0041]

The arylene group represented by L_1 include those with two or more arylene groups connected to each other via an alkylene group, -O-, -S- or -NR-, wherein R is an alkyl group, e.g., methyl or ethyl, or aryl group, e.g., phenyl, the latter being more preferable. The aryl groups include, in addition to phenyl,

A₁, A₂, and phenyl substituted with A₁ or A₂. The preferable alkylene groups include methylene and ethylene. These arylene groups are specifically described below:

[0042]

[Formula 9]

[0043]

Next, the formula (A-2) is described. In the formula (A-2), R₅₄ is the same as R₅₁ or R₅₂ in the formula (A-1), R₅₅ as R₅₃ in the formula (A-1), p₄ as p₁ or p₂ in the formula (A-1), and L₂ to L₁ in the formula (A-1). Their preferable examples are also similar.

[0044]

In the formula (A-2), p₅ is an integer of 0 to 4, particularly preferably 0 to 2. When p₅ is an integer of 1 to 3, particularly 1 or 2, each of R₅₅'s is preferably methyl or phenyl.

[0045]

In the formula (A-2), R₅₄ and R₅₅ may be the same or different. When there are two or more R₅₄'s, they may be the same or different, and so may be R₅₅'s.

[0046]

In the formula (A-1), at least one of Ar₁ and Ar₂, and at least one of Ar₃ and Ar₄ are preferably phenyl, biphenyl,

terphenyl, styryl, phenylstyryl, diphenylstyryl, thienyl, methylthienyl, phenylthienyl or phenylbithienyl. Moreover, at least one of Ar₁ and Ar₂, and at least one of Ar₃ and Ar₄ are preferably phenyl, biphenyl or terphenyl, and L₁ is preferably a single bond.

[0047]

In the formula (A-2), at least one of Ar₅ and Ar₆ is preferably phenyl, biphenyl, terphenyl, styryl, phenylstyryl, diphenylstyryl, thienyl, methylthienyl, phenylthienyl or phenylbithienyl. Moreover, at least one of Ar₅ and Ar₆ is preferably phenyl, biphenyl or terphenyl, and L₂ is preferably a single bond.

[0048]

The compounds represented by the formulae (A-1) and (A-2) are specifically described below, but the present invention is not limited by these compounds. These compounds are represented by a combination of formula and groups, where only substituents are described, when summarized by, e.g., R₃₂₋₃₇, and as -H when each of them is hydrogen atom.

[0049] [Formula 10] to [0078] [Formula 39]

[0079]

The methods of producing the phenyl anthracene derivatives useful for the present invention are described

elsewhere, e.g., Japanese Patent Laid-open No. 12600/1996.

[0080]

These compounds may be used either individually or in combination.

[0081]

The blue-emitting layer with a phenyl anthracene derivative as the blue-emitting compound is preferably 1 to 500nm thick, more preferably 10 to 200nm thick.

[0082]

The light-emitting layer may be doped with a dopant in such a way to keep the blue-emitting characteristics. The dopants useful for the present invention include styryl-based amine compounds or the like, disclosed by WO98/08360 and Japanese Patent Laid-open No. 239655/1996. These compounds are described in detail later. The dopant is preferably contained in the light-emitting layer at 0.1 to 20% by weight. Use of the dopant improves light-emitting efficiency and element stability.

[0083]

The blue-emitting layer may contain, as the host compound(s), a hole injecting/transferring compound and/or electron injecting/transferring compound in the hole transferring layer and electron transferring layer adjacent to the light-emitting layer, respectively. More specifically, a phenyl anthracene derivative for the electron transferring

layer may be used as the host compound. This compound, having the blue-emitting characteristics, can by itself emit blue color. However, when the host compound lacks such characteristics, it may be doped with a dopant to change its light-emitting characteristics in such a way that it can emit blue color. The styryl-based amine compound described above is one example of such a dopant.

[0084]

It is preferable to set the thickness ratio of the light-emitting layer to the electron or hole transferring layer containing the compound which serves as the host material at 1/100 to 100/1.

[0085]

The blue-emitting layer may be a mixed layer of hole injecting/transferring and electron injecting/transferring compounds, and such an embodiment is preferable. In particular, one of the hole injecting/transferring and electron injecting/transferring compounds is preferably the same as the one for the hole or electron transferring layer adjacent to the light-emitting layer. It is more preferable that the hole and electron transferring layers are provided adjacent to the light-emitting layer, and the hole injecting/transferring and electron injecting/transferring compounds for these layers are used for the mixed layer.

[0086]

More specifically, it is preferable to use the phenyl anthracene derivative in the electron transferring layer as the electron injecting/transferring compound, and the aromatic tertiary amine in the hole transferring layer as the hole injecting/transferring compound. The compound represented by the formula (A) is the preferable phenyl anthracene derivative, and the tetraaryl benzidine represented by the formula (1) is the preferable aromatic tertiary amine.

[0087]

[Formula 40]

(1)

[0088]

The formula (1) is described. R_1 to R_4 are each an aryl, alkyl, alkoxy, aryloxy or halogen group, and may be the same or different; r_1 to r_4 are each an integer of 0 to 5, when r_1 to r_4 are each an integer of 2 or more, adjacent R_1 's may be bonded to each other to form a ring, and so may be R_2 's, R_3 's and R_4 's; R_5 and R_6 are each an alkyl, alkoxy, amino or halogen group, and may be the same or different; and r_5 and r_6 are each an integer of 0 to 4.

[0089]

The aryl groups represented by R_1 to R_4 may be single- or poly-nuclear, including condensed and combined rings, preferably having a total carbon number of 6 to 20. They may

be substituted, and the preferable substituents include alkyl, alkoxy, aryl, aryloxy and amino groups, and halogen atom. More specifically, they include phenyl, o-, m- or p-tolyl, pyrenyl, perylenyl, coronenyl, naphthyl, anthryl, biphenyl, phenylanthryl and tolylanthryl groups, of which phenyl group is particularly preferable. It is also preferable that the aryl group, in particular phenyl group, is bonded to the 3-site (meta-site with respect to the site at which N is bonded) or 4-site (para-site with respect to the site at which N is bonded).

[0090]

The alkyl groups represented by R_1 to R_4 may be of straight-chain or branched, preferably having 1 to 10 carbon atoms. They may be substituted with a substituent similar to an aryl group, e.g., methyl, ethyl, n- or i- propyl, or n-, i-, s- or t-butyl group.

[0091]

The alkoxy groups represented by R_1 to R_4 preferably have 1 to 6 carbon atoms in the alkyl portion. More specifically, they include methoxy, ethoxy and t-butoxy. The alkoxy group may be substituted.

[0092]

The aryloxy groups represented by R_1 to R_4 include phenoxy, 4-methylphenoxy and 4-(t-butyl)phenoxy.

[0093]

The halogen groups represented by R_1 to R_4 include chlorine

and bromine atom.

[0094]

The preferred embodiments of the compounds represented by the formula (1) are those with at least one of r_1 to r_4 being an integer of 2 or more, where R_1 's are bonded to each other to form a ring (e.g., benzene ring), and so are R_2 's, R_3 's and R_4 's.

[0095]

Other preferred embodiments are those with at least one of R_1 to R_4 being an aryl group. In other words, r_1 to r_4 are not simultaneously zero, or $r_1 + r_2 + r_3 + r_4$ is an integer of 1 or more, which satisfies that at least one aryl group is present.

[0096]

When at least one of R_1 to R_4 is an aryl group, it is particularly preferable that 2 to 4 aryl groups are present as R_1 to R_4 in the molecule, and 2 to 4 of r_1 to r_4 are an integer of 1 or more. In particular, preferably a total of 2 to 4 aryl groups are present in the molecule, more preferably 2 to 4 of r_1 to r_4 are preferably an integer of 1, and still more preferably all of R_1 to R_4 are an integer of 1 and an aryl group. In other words, the four benzene rings in the molecule which may be substituted with R_1 to R_4 preferably have a total of 2 to 4 aryl groups, where the 2 to 4 aryl groups may be bonded to the same benzene ring or different benzene rings, more preferably the 2 to 4 aryl groups are bonded to the benzene rings which are

different from each other. Still more preferably, at least 2 aryl groups are bonded to the para-or meta-site with respect to the site at which N is bonded, where at least one of the aryl groups is preferably phenyl, i.e., the aryl group is bonded to the benzene ring at the 4- or 3-site with respect to N atom, to form the biphenyl group. In particular, 2 to 4 of the aryl groups preferably form 4- or 3-biphenyl groups. The 4- and 3-biphenyl groups may be present either individually or in combination. The specific preferable aryl groups other than phenyl include (1-,2-)naphthyl, (1-,2-,9-)anthryl, pyrenyl, perylenyl and coronenyl. They are preferably bonded to the para- or meta-site with respect to N atom, as is the case with phenyl group. Each of these may be present together with phenyl group.

[0097]

In the formula (1), the alkyl and alkoxy group and halogen atom represented by R_5 or R_6 may be the same as those represented by R_1 to R_4 .

[0098]

The amino groups represented by R_5 or R_6 may be substituted or not substituted, the former being more preferable. More specifically, the preferable ones include dimethylamino, diethylamino, diphenylamino, ditolylamino, dibiphenylamino, N-phenyl-N-tolylamino, N-phenyl-N-naphthylamino, N-phenyl-N-biphenylamino, N-phenyl-N-anthrylamino,

N-phenyl-N-pyrenylamino, dinaphthylamino, dianthrylamino and dipyrenylamino group.

[0099]

It is preferable that each of r_5 and r_6 is zero, and the biphenylene group with 2 arylamino groups bonded to each other is preferably not substituted.

[0100]

When r_1 to r_4 are each an integer of 2 or more, R_1 's may be the same or different, and so may be R_2 's to R_4 's. Similarly, when r_5 and r_6 are each an integer of 2 or more, R_5 's may be the same or different, and so may be R_6 's.

[0101]

Of these compounds, those represented by the following formula (1-1) are more preferable.

[0102]

[Formula 41]

(1-1)

[0103]

The formula (1-1) is described. A_{11} to A_{14} are each phenyl group or hydrogen atom bonded to the para-site (4-site) or meta-site (3-site) with respect to the site at which N is bonded. They may be the same or different. However, 2 or more of A_{11} to A_{14} are phenyl. Each phenyl group may be substituted, where the substituent may be the same as that for the aryl group

represented by R_1 to R_4 .

[0104]

R_7 to R_{10} are each an alkyl, alkoxy, aryl, aryloxy or halogen group, and may be the same or different. More specifically, they may be the same as those represented by R_1 to R_4 in the formula (1).

[0105]

r_7 to r_{10} are each an integer of 0 to 4, preferably 0.

[0106]

When r_7 to r_{10} are each an integer of 2 or more, R_7 's may be the same or different, and so may be R_8 's to R_{10} 's.

[0107]

R_5 and R_6 in the formula (1-1) are the same as those in the formula (1), and so are r_5 and r_6 , preferably $r_5 = r_6 = 0$.

[0108]

The compounds represented by the formula (1) are specifically described below, but the present invention is not limited by these compounds. These compounds are represented in the same manner as in the formulae (I) and (II), where R_1 to R_4 or the like are described by substituents, or H when all of them are H. These include N,N'-di(1-naphthyl)-N,N'-diphenylbenzidine, which was used in EXAMPLE.

[0109] [Formula 42] to [0114] [Formula 47]

[0115]

The tetraarylbenzidine derivatives represented by the formula (1) may be used either individually or in combination.

[0116]

The mixing ratio (by volume) of the electron injecting/transferring compound to hole injecting/transferring compound in the mixed layer is preferably 10/90 to 90/10, more preferably 20/80 to 80/20.

[0117]

When the phenyl anthracene derivative described earlier is used as the electron transferring compound in the mixed layer, it itself may serve as the blue-emitting compound. When the blue-emitting layer is composed of the phenyl anthracene derivative as the blue-emitting compound and tetraarylbenzidine derivative, the phenyl anthracene derivative/ tetraarylbenzidine derivative ratio (by volume) is preferably 95/5 to 30/70, more preferably 90/10 to 40/60.

[0118]

The above mixed layer may be doped with a dopant. Use of the dopant is preferable for improving light-emitting efficiency and element stability. The dopant is contained in the mixed layer at 0.1 to 20% by weight.

[0119]

The styryl-based amine compound is a preferable example

of the dopant. In particular, the compound represented by the formula (S) is preferable.

[0120]

(S)

[0121]

The formula (S) is described. R_{61} in the formula (S) is hydrogen or an aryl group. The aryl groups represented by R_{61} may be substituted, preferably having a total of 6 to 30 carbon atoms. Phenyl group is one example.

[0122]

R_{62} and R_{63} are each hydrogen, or an aryl or alkenyl group, and may be the same or different.

[0123]

The aryl groups represented by R_{62} or R_{63} may be substituted, particularly preferably having a total of 6 to 70 carbon atoms. More specifically, they include phenyl, naphthyl and anthryl, and the preferable substituents include arylamino (e.g., diphenylamino) and arylaminoaryl. These substituents are also preferably substituted with, e.g., styryl group (styryl, or phenyl, diphenylamino, naphthyl(phenyl)amino or diphenylaminophenyl). In this case, it is also preferable that the group has a structure with monovalent groups derived from the compounds represented by the formula (S) being bonded to each other directly or via a coupling group.

[0124]

The alkenyl groups represented by R_{62} or R_{63} may be substituted, preferably having a total of 2 to 50 carbon atoms. Vinyl group is one example. It preferably forms styryl group with vinyl group, and styryl group may be substituted with an arylaminoaryl group (e.g., diphenylaminophenyl group) or arylamino group (e.g., diphenylamino group). In this case, it is also preferable that the group has a structure with monovalent groups derived from the compounds represented by the formula (S) being bonded to each other directly or via a coupling group.

[0125]

R_{64} and R_{65} represent an arylamino or arylaminoaryl group, which may include styryl group (styryl group or further phenyl group as a substituent). In this case, it is also preferable that the group has a structure with monovalent groups derived from the compounds represented by the formula (S) being bonded to each other directly or via a coupling group, as is the case with the alkenyl groups described above.

[0126]

v_1 and v_2 are each an integer of 0 to 5. When v_1 is an integer of 2 or more, R_{64} 's may be bonded to each other to form a condensed ring, e.g., condensed benzene ring, and so may be R_{65} 's.

[0127]

R_{66} and R_{67} represent an alkyl or aryl group. The alkyl groups represented by R_{66} or R_{67} may be substituted, of straight-chain or branched, and preferably having a total of 1 to 6 carbon atoms. More specifically, they include methyl and ethyl. The aryl groups represented by R_{66} or R_{67} may be substituted, single- or poly-nuclear, and preferably having a total of 6 to 20 carbon atoms. More specifically, they include phenyl.

[0128]

v_3 and v_4 are each an integer of 0 to 4.

[0129]

v_5 is 0 or 1. Of the compounds represented by the formula (S), the more preferable ones have a structure in which v_5 is zero, and diphenyl amino group to which R_{64} and R_{65} may be bonded is at the para-site with vinyl group to which R_{61} , R_{62} and R_{63} are bonded in the phenylene group.

[0130]

The compounds represented by the following formula (S-1) or (S-2) are particularly preferable.

[0131]

[Formula 49]

(S-1)

(S-2)

[0132]

R_{61} , R_{62} , R_{64} , R_{65} , v_1 , and v_2 in the formula (S-1) are the same as the corresponding ones in the formula (S); n_1 is 0 or 1; and L_{61} is a bond or an arylene group. The preferable arylene groups include phenylene, biphenylene, naphthylene and anthraylene group, and a combination thereof is also preferable. They may be substituted.

[0133]

R_{61} to R_{63} , R_{65} and v_2 in the formula (S-2) are the same as the corresponding ones in the formula (S); n_2 is 0 or 1; and L_{62} is the same as L_{61} in the formula (S-1).

[0134]

The specific examples of the styryl-based amine compounds represented by the formula (S) are described below.

[0135]

[Formula 50]

[0136]

[Formula 51]

[0137]

These compounds may be used either individually or in combination.

[0138]

It is preferable to select the electron injecting/transferring and hole injecting/transferring

compounds for the mixed layer in such a way that they have almost the same product of charge mobility and charge density. More preferably, they satisfy the above conditions and have almost the same charge mobility. Charge mobility is preferably in a range of 1×10^{-1} to $1 \times 10^{-5} \text{ cm}^2/\text{V}\cdot\text{s}$, determined by the time of flight method. Selection of these compounds which have charge mobility levels close to each other brings about the advantages of (i) improved recombination probability of the carrier, which improves light-emitting efficiency, and (ii) reduced leakage of the carrier through the light-emitting layer, which reduces damages of the carrier transferring layer and extends light-emitting life of the element. Mixing the hole injecting/transferring and electron injecting/transferring compounds with each other brings the advantages, e.g., reduced charge mobility of the hole and electron, which improves the recombination probability.

[0139]

The electron injecting/transferring and hole injecting/transferring compounds may be mixed with each other for the mixed layer in such a way that each has a constant concentration in the layer, or to have a concentration distribution in the film thickness direction, the hole injecting/transferring compound concentration being higher on the hole transferring layer side and gradually decreasing towards the electron transferring layer, and so is vice versa

the electron injecting/transferring compound concentration being higher on the electron transferring layer side and gradually decreasing towards the hole transferring layer. It is preferable that around 95 to 50% by weight of the electron injecting/transferring compound is concentrated in the half of the mixed layer of inclined concentration on the electron transferring layer side, and so is vice versa for the hole injecting/transferring compound.

[0140]

The blue-emitting layer composed of the above mixed layer has the electrons and holes distributed throughout the light-emitting layer, and emits light not only from the vicinity of the interface between the layers but also from the mixed layer as a whole, because the recombination and light-emitting points are present throughout the mixed layer. This can be easily confirmed by fitting the spectral patterns of the observed emitted light with the spectral patterns obtained by simulating the optical interference between the reflected and direct light at the optical interface after supposing the light-emitting region. The layer which can emit light from its whole body brings advantages, e.g., stably emitting light of different wavelengths from the laminate, and extended service life of the light-emitting element.

[0141]

The maximum wavelength of light emitted from the

blue-emitting layer for the present invention is 400 to 500nm.

[0142]

The above-described mixed layer has a thickness of 1 to 500nm, preferably 20 to 200nm.

[0143]

<Other Emitted Light Colors>

The organic EL element of the present invention preferably includes, in addition to the blue-emitting layer, at least one layer emitting light of wavelength different from that of blue, to cope with the requirements of emitting multi-colors. Such a layer may emit red color (maximum wavelength of emitted light: 600 to 700nm) or green color (maximum wavelength of emitted light: 500 to 560nm).

[0144]

These light-emitting layers are preferably composed of the mixed layer which contains the same host compound as the blue-emitting layer and is doped with a dopant to emit light other than blue, because this extends the recombination region and provides the favorable conditions for generating excitons.

[0145]

One of the preferred embodiments of the mixed layer contains a mixture of the above-described phenyl anthracene derivative and tetraarylbenzidine doped with a naphthacene derivative. For example, when rebrene is used as the naphthacene derivative, the mixed layer can emit red color

(maximum wavelength of emitted light: 540 to 600nm). Addition of the naphthacene derivative is preferable for extending service life of the element. Apentacene derivative also brings the similar advantage. These compounds are described in, e.g., Japanese Patent Laid-Open No. 311442/1996, WO98/08360 and Japanese Patent Application No. 137505/1998.

[0146]

The preferable naphthacene compounds are those represented by the formula (N):

[0147]

[Formula 52]

(N)

[0148]

wherein, R_a , R_b , R_c and R_d are each an alkyl, aryl, amino, heterocyclic or alkenyl group, which may be substituted or not substituted, of which an aryl, amino, heterocyclic and alkenyl are more preferable.

[0149]

The aryl groups represented by R_a , R_b , R_c or R_d may be single- or poly-nuclear, including condensed and combined rings, preferably having a total carbon number of 6 to 30. They may be substituted.

[0150]

More specifically, the preferable aryl groups represented

by R_a , R_b , R_c or R_d include phenyl, o-, m- or p-tolyl, pyrenyl, perylenyl, coronenyl, (1-,2-)naphthyl, anthryl, o-, m- or p-biphenyl, terphenyl and phenylanthryl.

[0151]

The amino groups represented by R_a , R_b , R_c or R_d include alkyl amino, aryl amino and aralkyl amino. They preferably have an aliphatic ring having a total of 1 to 6 carbon atoms and/or aromatic ring having a total of 1 to 4 carbon atoms. More specifically, they include dimethylamino, diethylamino, dibutylamino, diphenylamino, ditolylamino, bisdiphenylamino and bisnaphthylamino.

[0152]

The heterocyclic groups represented by R_a , R_b , R_c or R_d include 5- and 6-membered aromatic heterocyclic groups with O, N or S as the heteroatom, and condensed polynuclear heterocyclic groups of 2 to 20 carbon atoms. More specifically, the aromatic heterocyclic and condensed polynuclear heterocyclic groups include thienyl, furyl, pyrrolyl, pyridyl, quinolyl, quinoxalyl.

[0153]

The preferable alkenyl groups represented by R_a , R_b , R_c or R_d include (1- and 2-)phenylalkenyl, (1,2- and 2,2-)diphenylalkenyl and (1,2,2-)triphenylalkenyl, which have at least one phenyl group as the substituent.

However, they may not be substituted.

[0154]

When each of R_a , R_b , R_c and R_d has a substituent(s), at least 2 of them are preferably aryl, amino, heterocyclic, alkenyl or aryloxy. The aryl, amino, heterocyclic and alkenyl groups are similar to those for R_a , R_b , R_c and R_d .

[0155]

The aryloxy group for these substituents preferably has an aryl group of 6 to 18 carbon atoms. These include (o-, m- and p-)phenoxy.

[0156]

Two or more of these substituents may form a condensed ring. Moreover, these substituents may be further substituted, where the preferable substituents are similar to those described above.

[0157]

When each of R_a , R_b , R_c and R_d has a substituent(s), at least 2 of them are preferably substituted at a position not limited. In other words, they may be substituted at meta, para or ortho site. R_a and R_d are preferably the same, although may not be the same, and so are R_b and R_c .

[0158]

R_e , R_f , R_g and R_h are each hydrogen atom, or alkyl, aryl, amino or alkenyl which may be substituted.

[0158]

The alkyl groups represented by R_e , R_f , R_g or R_h preferably

have 1 to 6 carbon atoms, and may be of straight-chain or branched. The preferable alkyl groups include methyl, ethyl, (n- or i-)propyl, (n-, i-, sec- or tert-)butyl and (n-, i-, neo- or tert-)pentyl.

[0160]

The aryl, amino and alkenyl groups represented by R_e , R_f , R_g or R_h are similar to those for R_a , R_b , R_c and R_d . R_e and R_f are preferably the same, although may not be the same, and so are R_g and R_h .

[0161]

The specific examples of the naphthacene derivatives are described below. These examples are described by the combinations shown in the formula (N).

[0162] [Formula 53] to [0165] [Formula 56]

[0166]

These compounds may be used either individually or in combination.

[0167]

The naphthacene derivative is contained in the mixed layer preferably at 0.1 to 20% by weight.

[0168]

The phenyl anthracene derivative/tetraarylbenzidine ratio in the mixed layer is preferably 90/10 to 10/90 by volume. The mixed layer is preferably 1 to 500nm thick, more preferably 10 to 200nm.

[0169]

The present invention may be structured to have 2 or 3 light-emitting layers, including the blue-emitting layer, to emit white-colored light.

[0170]

<Hole transferring and/or injecting layers>

The present invention is preferably provided with the hole transferring and/or injecting layers, as partly described earlier. The hole transferring and/or injecting layers (sometimes referred to as the hole injecting/transferring layer) is preferably provided, even in the case of an embodiment in which the hole transferring/injecting compound is not serving as the host material of the light-emitting layer in the element provided with the hole transferring layer. The hole injecting/transferring compound is preferably an aromatic tertiary amine. The preferable amines include the tetraarylbenzidine derivative represented by the formula (1) and triphenylamine derivative represented by the formula (2). The formula (1) is described earlier. The formula (2) is described below.

[0171]

[Formula 57]

(2)

[0172]

wherein, two ϕ are phenylene groups, and the biphenylene groups

represented by ϕ - ϕ include 4,4'-biphenylene, 3,3'-biphenylene, 3,4'-biphenylene, 2,2'-biphenylene, 2,3'-biphenylene and 2,4'-biphenylene, of which 4,4'-biphenylene is more preferable.

[0173]

R_{01} , R_{02} , R_{03} and R_{04} are each an alkyl, aryl or diarylaminoaryl group,

[0174]

[Formula 58]

(a-1), (a-2) or (a-3)

wherein, R_{011} , R_{012} , R_{013} , R_{014} , R_{015} , R_{016} and R_{017} are each an aryl group;

[0175]

and may be the same or different. However, at least one of R_{01} to R_{04} is diarylaminoaryl group, or one of (a-1) to (a-3). The aryl group represented by R_{011} , R_{012} , R_{013} , R_{014} , R_{015} , R_{016} or R_{017} may be substituted or not substituted.

[0176]

The alkyl groups represented by R_{01} , R_{02} , R_{03} or R_{04} are may be substituted, and of straight-chain or branched, preferably having 1 to 20 carbon atoms. More specifically, they include methyl and ethyl.

[0177]

The aryl groups represented by R_{01} , R_{02} , R_{03} , R_{04} , R_{011} , R_{012} , R_{013} , R_{014} , R_{015} , R_{016} or R_{017} may be single- or poly-nuclear,

preferably having a total carbon number of 6 to 20. More specifically, they include phenyl, naphthyl, anthryl, phenylanthryl, pyrenyl, perylenyl, and o-, m- or p-biphenyl. These aryl groups may be substituted, and the substituents include alkyl, aryl and alkoxy which are substituted or not substituted, aryloxy and $-N(R_{021})R_{022}$ of 1 to 6 carbon atoms, wherein R_{021} and R_{022} are each an aryl group which is substituted or not substituted.

[0178]

The aryl groups represented by R_{021} or R_{022} may be single- or poly-nuclear, preferably having a total carbon number of 6 to 20. More specifically, they include phenyl, naphthyl, anthryl, phenylanthryl, pyrenyl, perylenyl, and o-, m- or p-biphenyl, of which phenyl is more preferable. These aryl groups may be substituted, and the substituents include alkyl and aryl which is substituted or not substituted of 1 to 6 carbon atoms. The preferable alkyl group is methyl, and preferable aryl group is phenyl.

[0179]

The diarylaminoaryl groups represented by R_{01} , R_{02} , R_{03} or R_{04} include diarylaminophenyl, and the diarylamino group in the diarylaminoaryl group is preferably bonded at the meta-site (3-site) or para-site (4-site) with respect to the structure represented by the formula (2). The phenyl group may be substituted, but preferably only with diarylamino group.

[0180]

The aryl group in the diarylamino group may be single- or poly-nuclear, preferably having a total carbon number of 6 to 20. More specifically, they include phenyl, naphthyl, anthryl, phenylanthryl, pyrenyl, perylenyl, and o-, m- or p-biphenyl, of which phenyl is more preferable. These aryl groups may be substituted, and the substituents include alkyl and aryl which is substituted or not substituted of 1 to 6 carbon atoms. The preferable alkyl group is methyl, and preferable aryl group is phenyl. The preferable substituents for the aryl group include those described above except the diarylaminoaryl groups represented by R_{01} to R_{04} in the formula (2). When there are 2 or more substituents, they may be the same or different. The substituent is preferably bonded to the meta- or para-site with respect to the site at which N is bonded.

[0181]

In the formula (2), r_{01} , r_{02} , r_{03} and r_{04} are each an integer of 0 to 5, preferably 0 to 2, more preferably 0 or 1. Moreover, $r_{01}+r_{02}+r_{03}+r_{04}$ is 1 or more, preferably 1 to 4, more preferably 2 to 4. R_{01} , R_{02} , R_{03} or R_{04} is bonded to the meta- or para-site with respect to the site at which N is bonded. R_{01} , R_{02} , R_{03} and R_{04} may be bonded totally to the meta- or para-site, or some may be bonded to the meta-site and others to the para-site. When r_{01} , r_{02} , r_{03} or r_{04} is each an integer of 2 or more, R_{01} 's may be the same or different and the adjacent ones may be bonded

to each other to form a ring, and so may be R_{02} 's, R_{03} 's and R_{04} 's. The ring may be aromatic, e.g., benzene ring, or aliphatic, e.g., cyclohexane ring.

[0182]

Described below are some of the preferable compounds represented by the formula (2), although not limited thereto.

[0183]

[Formula 59]

[0184]

[Formula 60]

[0185]

They may be used either individually or in combination.

[0186]

When the hole transferring and injecting layers are provided in this order from the light-emitting layer, it is preferable to use the compound represented by the formula (1) for the former, and the one represented by the formula (2) for the latter. This combination of the compounds improves the function of blocking the electrons. In any case, it is preferable that the hole transferring layer has the benzidine structure and an aromatic tertiary amine lacking the phenylenediamine structure, and the hole injecting layer has an aromatic tertiary amine with the phenylenediamine structure.

[0187]

The hole injecting layer is 1 to 1000nm thick, preferably 1 to 100nm thick, and the hole transferring layer is 1 to 200nm thick, preferably 5 to 100nm thick. When only one layer having the hole injecting and transferring functions is provided, it is 1 to 1000nm thick, preferably 10 to 500nm thick.

[0188]

<Electron transferring and/or injecting layers>

The present invention is preferably provided with the electron transferring and/or injecting layers, as partly described earlier. The electron transferring and/or injecting layers (sometimes referred to as the electron injecting/transferring layer) is preferably provided, even in the case of an embodiment in which the electron transferring/injecting compound is not serving as the host material of the light-emitting layer in the element provided with the electron transferring layer. The examples of the electron injecting/transferring compounds include, in addition to the phenylanthracene derivatives described earlier, organometallic complexes with 8-quinolinol or its derivative (e.g., tris(8-quinolinolate)aluminum (AlQ3)) as the ligand, e.g., the quinoline, oxadiazole, perylene, pyridine, pyrimidine, quinoxaline, diphenylquinone and nitro-substituted fluorine derivatives.

[0189]

It is particularly preferable to use the diphenylanthracene derivative represented by the formula (A) and an aluminum complex with 8-quinolinol or its derivative (e.g., tris(8-quinolinolate)aluminum (AlQ_3)) as the ligand, the former for the electron transferring layer on the light-emitting layer side and the latter for the electron injecting layer on the cathode side. The aluminum complexes with 8-quinolinol or its derivative as the ligand are described in WO98/08360.

[0190]

The electron injecting layer is 1 to 1000nm thick, preferably 1 to 100nm thick, and the electron transferring layer is 1 to 500nm thick, preferably 1 to 100nm thick. When only one layer having the electron injecting and transferring functions is provided, it is 1 to 1000nm thick, preferably 1 to 100nm thick.

[0191]

<Cathode>

The preferable cathode materials for the present invention include halides and oxides of alkali metals, e.g., Li, Na, K, Rb and Cs). More specifically, they include the halides, e.g., lithium fluoride (LiF), lithium chloride (LiCl), lithium bromide (LiBr), lithium iodide (LiI), sodium fluoride (NaF), sodium chloride (NaCl), sodium bromide (NaBr), sodium iodide (NaI), rubidium fluoride (RbF), rubidium chloride (RbCl),

rubidium bromide (RbBr), rubidium iodide (RbI), cesium fluoride (CsF), cesium chloride (CsCl), cesium bromide (CsBr), cesium iodide (CsI); and oxides, e.g., lithium oxide (Li_2O) and sodium oxide (Na_2O), of which halides of Rb and Cs are more preferable, and in particular chlorides and iodides are still more preferable.

[0192]

The halide or oxide of an alkali metal as the undercoat may be coated with a material of low work function (e.g., Li, Na, K, Mg, Al, Ag, In, or an alloy containing one or more of these metals), to form the laminate. The cathode is preferably composed of fine grains, and more preferably of amorphous grains. The cathode preferably has a total thickness of 10 to 1000nm. The undercoat layer, when provided, is around 0.1 to 1nm thick.

[0193]

Use of a halide or oxide of an alkali metal as the material for the cathode is particularly useful for the element having a blue-emitting layer, because it can help the element stably emit blue color. The host of a blue-emitting system has a larger energy gap than that of a green-emitting device, and hence needs electron and hole injecting capacities of higher efficiency. The conventional cathode of MgAg is insufficient in electron injecting efficiency, and an alkali metal is a desirable substitute to improve the efficiency, because of its lower work function. It keeps its work function unchanged even when

transformed into the halide or oxide, and can be returned back to the metallic state by reduction or the like when placed in an electric field. Therefore, it is an optimum electron injecting material which can be easily handled. It also works to improve adhesion between the organic film and electrode.

[0194]

Use of a halide or oxide of an alkali metal for the cathode is essential particularly for the embodiment in which the blue-emitting layer contains no electron or hole injecting/transferring compound as the host material, which is contained in the electron or hole transfer layer as the adjacent layer.

[0195]

The organic layer at the interface with the cathode may be doped with a metal, e.g., Li.

[0196]

Coating the electrode with Al- or F-based compound by evaporation or sputtering in the final stage of the production process improves its sealing effect.

[0197]

In an embodiment in which tris(8-quinolinolate)aluminum (AlQ3) or the like is used for the electron injecting and/or transferring layers and cathode is formed by sputtering, a 0.1 to 20nm thick layer of a naphthacene derivative (described earlier), e.g., rubrene, may be formed between the electron

injecting or transferring layer and cathode, to prevent damages of the electron injecting or transferring layer by sputtering.

[0198]

<Anode>

For an organic EL element to emit light from the whole surface, at least one of its electrodes must be transparent or semi-transparent. This limits the materials for the cathode. It is therefore preferable to determine the anode material and thickness in such a way to keep transmittance of emitted light preferably at 80% or more. More specifically, the anode is preferably of ITO (tin-doped indium oxide), IZO (zinc-doped indium oxide), SnO_2 , Ni, Au, Pt, Pd or dopant-containing polypyrrole, of which ITO or IZO is particularly preferable. ITO generally contains In_2O_3 and SnO in the stoichiometric ratio, but the oxygen content may be slightly deviated from the stoichiometric level. IZO generally contains In_2O_3 and ZnO in the stoichiometric ratio, but the oxygen content may be slightly deviated from the stoichiometric level. In ITO, the $\text{SnO}_2/\text{In}_2\text{O}_3$ ratio is 1 to 20% by weight, preferably 5 to 12%. In IZO, the $\text{ZnO}/\text{In}_2\text{O}_3$ ratio is normally around 12 to 32% by weight. The anode is preferably 10 to 500nm thick. Reliability of the element increases as the driving voltage decreases. It is preferably 10 to 30 Ω/\square or 10 Ω/\square or less (normally 0.1 to 10 Ω/\square) for ITO.

[0199]

For a large-sized device, e.g., display, an Al wiring system may be adopted to compensate for increased resistance of ITO.

[0200]

<Substrate materials>

The substrate materials are not limited. However, they are transparent or semi-transparent, e.g., glass or resin, to fetch emitted light from the substrate side. A color filter film, fluorescence conversion filter film or dielectric reflection film may be used for the substrate. Moreover, the substrate itself may be colored to control emitted light color.

[0201]

The color filter for liquid crystal display or the like may be used for the present invention. It is recommended to optimize fetching efficiency and color purity by tuning up the characteristics of the color filter specifically for light emitted from the organic EL element.

[0202]

The element will have improved resistance to light and display contrast, when provided with the color filter capable of cutting external light of short wavelength which the EL element material or fluorescent conversion layer may absorb.

[0203]

The color filter may be replaced by a thin optical film, e.g., dielectric multi-layered film.

[0204]

The fluorescent conversion filter film converts color of light emitted from an EL device by absorbing the color and emitting it from the fluorescent material in the film. It is composed of three components of binder, fluorescent material and light-absorbing material.

[0205]

The fluorescent material basically should have a high fluorescence quantum yield, and preferably has a strong absorption in the wavelength region of EL-emitted light. In practice, the laser colors are suitable. The useful compounds include those based on rhodamine, perylene, cyanine, phthalocyanine (including sub-phthalocyanine), naphthaloimide, hydrocarbons with condensed rings, condensed heterocyclic compounds, styryl and coumarin.

[0206]

The binder is basically selected from the materials which prevent extinction of fluorescence. They are preferably formed into fine patterns by photolithography, printing or the like. They are also preferably prevented from being damaged while the ITO film is being produced.

[0207]

The light-absorbing material is used when the fluorescent material is insufficient in light-absorbing capacity. It is selected from the materials which prevent extinction of fluorescence by the fluorescent material.

[0208]

<Modulation of emitted light by color filter>

Use of a color filter allows to modulate light emitted from the organic EL element of the present invention, and thereby easily provides the multi-color emitting organic EL display (multi-color emitting organic EL element).

[0209]

When the organic EL element is applied to the above device, it is preferable that the element including the light-emitting layer is placed between electrodes facing each other, and that at least one of the electrodes is transparent. It is necessary for at least one of the electrodes to be transparent for the element which includes a color filter. When the element emits light from the transparent electrode side, the color filter is provided on that side.

[0210]

The organic layer means the layer which contains an organic compound, and the organic compounds useful for the organic layer include metallic complexes with an organic compound as the ligand, and organometallic compounds.

[0211]

The above device may have a segment type or dot matrix type display, or both.

[0212]

A dot matrix type display has a pair of XY matrix type

electrodes, each being composed of two or more electrodes intersecting with each other and being positioned to face each other, where an organic layer is placed between the electrodes intersecting with each other to form the picture element. The color filter is preferably provided on the transparent electrode side of the picture element. Moreover, it is preferable to provide a black matrix around the picture element and in the vicinity of the position at which the color filter is provided (normally between the color filters). The black matrix can prevent leakage of light between the color filters, and thereby enhance visibility of emitted multi-colors.

[0213]

The picture element means the region of image-displaying arrays which is excited independently from another region to emit light.

[0214]

The two or more electrodes described above are normally stripe-shaped, and a pair of the electrodes are arranged in such a way to intersect each other at an almost right angle. The stripe-shaped electrodes are arranged normally by forming one electrode first and then the other. At the same time, the dot matrix type displaying section is frequently formed using an interlayer insulation film. Therefore, the stripe-shaped electrodes formed afterward may not be arranged almost on the same plane, or one stripe in the same direction may not form

a continuous film, which, however, is acceptable so long as there is a section where the electrodes intersect each other at an almost right angle.

[0215]

The dot matrix type displaying section may be formed by, e.g., the following method. A transparent substrate (e.g., of glass) is coated with a given color filter layer, and then with a 1 μ m to 5mm thick overcoat layer of transparent resin (e.g., arylilic or polyimide resin) on the color filter layer surface on which the transparent electrode(s) is (are) to be formed, preferably to improve flatness of the surface. The overcoat layer also works as the protective layer for the color filter. The overcoat layer is patterned to form the transparent electrode(s) thereon. A transparent, electrically insulating inorganic oxide layer may be provided as the passivation film between the transparent electrode(s) and overcoat layer.

[0216]

The plane having the patterned transparent electrode layer is provided with a 10nm to 100 μ m thick interlayer insulation film to cover the plane except for the electrode layer. The insulation film may be of an inorganic compound (e.g., SiO₂ or SiN_x) or resin (e.g., polyimide, acrylic or epoxy resin).

[0217]

Moreover, the plane may be provided with a spacer, in

addition to the insulation film, on the insulation layer, and further with an overhung body wider than the spacer on the spacer, to divide the element (Japanese Patent Laid-open No. 330792/1997).

[0218]

Then, the organic layer which contains the light-emitting layer in the organic EL element is formed, and the counter electrode is provided in such a way to intersect the transparent electrode. This provides the intersection of the electrode pair with the function of emitting light. It is preferable to leave the insulation layer after the element is formed, because it works to avoid useless light emission in an area unseen from the substrate surface. When the black matrix is used, its layer may be provided between the color filter layers.

[0219]

<Color filter and black matrix>

1) Color filter

The color filters useful for the present invention include the solid of the following pigment or the pigment dissolved or dispersed in a binder resin.

[0220]

The following pigments are useful for the present invention. Red (R) pigments: those based on perylene, rake, azo, quinacridon, anthraquinone, anthracene, isoindoline and isoindolynone. They may be used either individually or in

combination.

[0221]

Green (G) pigments: those based on phthalocyanine and copper phthalocyanine substituted with 2 or more halogen atoms, triphenylmethane, isoindoline and isoindolynone. They may be used either individually or in combination.

[0222]

Blue (B) pigments: those based on copper phthalocyanine, indanthrone, indophenol, cyanine and dioxadine. They may be used either individually or in combination.

[0223]

On the other hand, the binder resin is preferably transparent (transmitting at least 50% of visible light). The transparent resins (high-molecular-weight compounds) useful for the present invention include polymethyl methacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinyl pyrrolidone, hydroxyethyl cellulose and carboxymethyl cellulose.

[0224]

A photosensitive resin to which photolithography is applicable may be selected, in order to two-dimensionally arrange the divided color filter. These resins include light-curable resist materials having a reactive vinyl group, e.g., those based on acrylic acid, methacrylic acid, polyvinyl cinnamate and cyclic rubber. When the printing method is used,

a printing ink (medium) comprising a transparent resin may be selected. These transparent resins include compositions of monomer, oligomer or polymer for polyvinyl chloride, melamine, phenol, alkyd, epoxy, polyurethane, polyester, maleate and polyamide resins; polymethyl methacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinyl pyrrolidone, hydroxyethyl cellulose and carboxymethyl cellulose.

[0225]

When the color filter is mainly composed of a pigment, the film is generally prepared by vacuum evaporation or sputtering via the mask with a desired color filter pattern. When the color filter is composed of a pigment and binder resin, on the other hand, the fluorescent pigment is generally mixed with, or dispersed or dissolved in, the resin and resist, and the resultant coating solution is spread by spin coating, roll coating, casting or the like to form the film, which is provided with a desired color filter pattern by photolithography, printing or the like, and then cured under heating.

[0226]

The color filters prepared by the above methods preferably have film thickness and transmittance in the following ranges: film thickness: 0.5 to 20 μ m and transmittance: 50% or more (610nm) for the R pigment; film thickness: 0.5 to 20 μ m and transmittance: 50% or more (545nm) for the G pigment; and film thickness: 0.2 to 20 μ m and transmittance: 50% or more (460nm)

for the B pigment.

[0227]

When the color filter is composed of the pigment and binder, in particular, the pigment concentration is set within the limits in which the color filter can be patterned without any difficulty and sufficiently transmit light emitted from the organic EL element. The pigment is contained in the color filter containing the binder at 5 to 50% by weight, although the adequate content varies depending on pigment type.

[0228]

ii) Black matrix

The black matrixes useful for the present invention include the following thin films of metals and metal oxides, and black pigments. More specifically, thin films of metals and metal oxides useful for the present invention include those of chromium (Cr), nickel (Ni), copper (Cu) and the oxides thereof. The mixture of the above metal or its oxide preferably has an optical concentration of 3.0 or more (film thickness: 10 to 30nm (100 to 3000Å)).

[0229]

The concrete examples of the black pigments include carbon, titanium and aniline black, a pigment mixed with the color filter pigment to be blackened, and solidified pigment after being dissolved or dispersed in the binder resin as is the case with the one for the color filter.

[0230]

The thin film of metal or its oxide is formed by sputtering, evaporation, CVD or the like on the entire surface of the insulating substrate or at least on the entire surface of the display section by masking, and patterned by photolithography to form the black matrix pattern.

[0231]

When the black pigment is used, the black matrix can be formed by patterning in the manner similar to that for the color filter.

[0232]

<Protective layer (transparent, flat film)>

The protective layer (transparent, flat film), which may be provided as required for the present invention, is used to protect the color filter (including the black matrix) from being physically damaged, or deteriorated by the external environmental factors (e.g., water, oxygen or light). The material therefor is preferably transparent (transmitting 50% or more of visible light).

[0233]

More specifically, the resins useful for the present invention include light-curable and/or thermosetting resins, e.g., acrylate- and methacrylate-based resin having a reactive vinyl group. They also include monomers, oligomers or polymers for the transparent resins, e.g., melamine, phenol, alkyd, epoxy,

polyurethane, polyester, maleate, and polyamide resins; and transparent resins, e.g., polymethyl methacrylate, polyacrylate, polycarbonate, polyvinyl alcohol, polyvinyl pyrrolidone, polyimide, hydroxyethyl cellulose and carboxymethyl cellulose. The protective layer may be incorporated with an ultraviolet absorber to enhance resistance of the color filter and organic EL element to light.

[0234]

The protective layer is formed by spin coating, roll coating, casting or the like, when the above material is liquid. The resulting film is irradiated with light and cured under heating when the film material is a light-curable resin, or directly cured under heating when it is a thermosetting resin. When the layer is film-shaped, it may be stuck to the color filter by an adhesive agent.

[0235]

The protective layer is not limited in thickness, because visual field angle is little affected by the thickness. However, it may decrease light transmittance, when excessively thick. Therefore, thickness can be selected normally from a range of $1\mu\text{m}$ to 5nm .

[0236]

<Transparent, electrically insulating inorganic oxide layer>

The transparent, electrically insulating inorganic oxide layer for the present invention may be formed on the color filter

or protective layer by, e.g., evaporation, sputtering, dipping or the like. The layer may be composed of a single layer or 2 or more layers. When it is composed of 2 layers, the upper inorganic oxide layer controls elution of the inorganic ion from the lower inorganic oxide layer (e.g., of soda soda-lime glass), thereby protecting the organic EL element from the eluted ion.

[0237]

The materials useful for the inorganic oxide layer include silicon oxide (SiO_2), aluminum oxide (Al_2O_3), titanium oxide (TiO_2), yttrium oxide (Y_2O_3), germanium oxide (GeO_2), zinc oxide (ZnO), magnesium oxide (MgO), calcium oxide (CaO), boron oxide (B_2O_3), strontium oxide (SrO), barium oxide (BaO), lead oxide (PbO), zirconia (ZrO_2), sodium oxide (Na_2O), lithium oxide (Li_2O) and potassium oxide (K_2O), of which silicon oxide, aluminum oxide and titanium oxide are more preferable, because their layers (films) are highly transparent, formed into a film at a relatively low temperature (250°C or lower), and little harmful to the color filter or protective layer.

[0238]

The transparent, electrically insulating inorganic oxide layer is more preferably of glass plate, or glass plate coated with a film of at least one compound selected from the group consisting of the above-described oxides, e.g., silicon oxide, aluminum oxide and titanium oxide, on at least one side of the

transparent, electrically insulating glass plate, because it can be handled at low temperature (150°C or lower) required only to stick it to the color filter or protective layer while causing any damage on the color filter or protective layer. A glass plate can very efficiently cut off gases which may cause deterioration of the element, in particular steam, oxygen, monomer or the like.

[0239]

The glass plate compositions given in Tables 1 and 2 can be cited as the examples. In particular, those which can be cited include soda-lime glass, glass containing barium or strontium, lead glass, aluminosilicate glass, borosilicate glass and barium borosilicate glass. The electrically insulating inorganic oxide layer is compositionally not limited, so long as it is mainly composed of an inorganic oxide. It may contain a nitride (e.g., Si_3N_4).

[0240]

Thickness of the transparent, electrically insulating inorganic oxide layer is not limited, so long as it has no adverse effect on the organic EL element for emitting light. However, it is preferably 0.01 μm or more but 200 μm or less. The glass plate, or glass plate coated with a film of at least one compound selected from the group consisting of the above-described oxides, e.g., silicon oxide, aluminum oxide and titanium oxide, on at least one side of the transparent, electrically insulating glass

plate preferably has a thickness of $1\mu\text{m}$ or more but $200\mu\text{m}$ or less, viewed from its precision and strength. The inorganic oxide layer nears the monolayer of the inorganic oxide particles as its thickness decreases, having difficulties in cutting off gases which may cause deterioration of the element, e.g., steam, oxygen, monomer or the like discharged from the color filter or protective layer. As its thickness increases, on the other hand, light emitted from the organic EL element may leak out of the gap with the color filter, narrowing visual field angle of the multi-colored emitted light and possibly deteriorating practicality of the multi-color emitting device, although the critical thickness depends on fineness of the color filter.

[0241]

[Table 1]

Glass composition systems	
1) $R_2O-R' O-SiO_2$ • $Na_2O-CaO/MgO-SiO_2$ (Soda-lime glass) • $Na_2O/K_2O-BaO/SrO-SiO_2$ • $Na_2O/K_2O-CaO/ZnO-SiO_2$	
2) $R_2O-PbO-SiO_2$ • $K_2O/Na_2O-PbO-SiO_2$ (Lead glass)	
3) $R_2O-B_2O_3-SiO_2$ • $Na_2O-B_2O_3-SiO_2$ (Borosilicate glass) • $K_2O-B_2O_3-SiO_2$	
4) $R' O-B_2O_3-SiO_2$ • $PbO-B_2O_3-SiO_2$ • $PbO/ZnO-B_2O_3-SiO_2$ • $PbO-B_2O_3-SiO_2+(Filler)$ • $ZnO-B_2O_3-SiO_2$	
5) $R' O-Al_2O_3-SiO_2$ • $CaO/MgO-Al_2O_3-SiO_2$ (Aluminosilicate glass) • $MgO-Al_2O_3-SiO_2$ • $PbO/ZnO-Al_2O_3-SiO_2$	
6) $R_2O-Al_2O_3-SiO_2$ • $Li_2O-Al_2O_3-SiO_2$ • $Na_2O-Al_2O_3-SiO_2$	
7) $R' O-TiO_2-SiO_2$ • $BaO-TiO_2-SiO_2$	
8) $R_2O-ZrO_2-SiO_2$ • $Na_2O/Li_2O-ZrO_2-SiO_2$	
9) $R' O-P_2O_5-SiO_2$ • $CaO-P_2O_5-SiO_2$	
10) $R' O-SiO_2$ • $CaO/BaO/PbO-SiO_2$	
11) SiO_2	
12) $R_2O-R' O-B_2O_3$ • $Li_2O-BeO-B_2O_3$	
13) $R' O-R_2' O_3-B_2O_3$ • $CaO/BaO-Al_2O_3-B_2O_3$ • $CaO/PbO-Lu_2O_3-B_2O_3$	

R : Monovalent element
R' : Divalent element
R'' : Trivalent element

14) $R_2O-Al_2O_3-P_2O_5$ • $K_2O-Al_2O_3-P_2O_5$
15) $R' O-Al_2O_3-P_2O_5$ • $BaO/CaO-Al_2O_3-P_2O_5$ • $ZnO-Al_2O_3-P_2O_5$

[0242]

[Table 2]

	Classification	Compositions (mainly represented by 1- to 3-component systems)
1	Simple oxide	SiO_2 , B_2O_3 , GeO_2 , As_2O_3
2	Silicate	Li_2O-SiO_2 , Na_2O-SiO_2 , K_2O-SiO_2 $MgO-SiO_2$, $CaO-SiO_2$, $BaO-SiO_2$, $PbO-SiO_2$ $Na_2O-CaO-SiO_2$ $Al_2O_3-SiO_2$
3	Borate	$Li_2O-B_2O_3$, $Na_2O-B_2O_3$, $K_2O-B_2O_3$ $MgO-B_2O_3$, $CaO-B_2O_3$, $PbO-B_2O_3$ $Na_2O-CaO-B_2O_3$, $ZnO-PbO-B_2O_3$ $Al_2O_3-B_2O_3$, $SiO_2-B_2O_3$
4	Phosphate	$Li_2O-P_2O_5$, $Na_2O-P_2O_5$ $MgO-P_2O_5$, $CaO-P_2O_5$, $BaO-P_2O_5$ $K_2O-BaO-P_2O_5$ $Al_2O_3-P_2O_5$, $SiO_2-P_2O_5$, $B_2O_3-P_2O_5$ $V_2O_5-P_2O_5$, $Fe_2O_3-P_2O_5$, $WO_3-P_2O_5$
5	Germanate	Li_2O-GeO_2 , Na_2O-GeO_2 , K_2O-GeO_2 , $B_2O_3-GeO_2$, SiO_2-GeO_2
6	Tungstate	Na_2O-WO_3 , K_2O-WO_3
7	Molybdate	Na_2O-MoO_3 , K_2O-MoO_3 , Li_2O-MoO_3
8	Tellurate	Na_2O-TeO_2
9	Borosilicate	$Na_2O-B_2O_3-SiO_2$
10	Aluminosilicate	$Na_2O-Al_2O_3-SiO_2$, $CaO-Al_2O_3-SiO_2$
11	Aluminoborate	$CaO-Al_2O_3-B_2O_3$, $ZnO-Al_2O_3-B_2O_3$,
12	Aluminoborosilicate	$Na_2O-Al_2O_3-B_2O_3-SiO_2$
13	Fluoride	BeF_2 , $NaF-BeF_2$ $ZrF_4-BaF_2-ThF_4$, $GdF_3-BaF_2-ZrF_4$
14	Fluorophosphate	$Al(PO_3)_3-AlF_3-NaF-CaF_2$
15	Oxyhalide	$Ag_2O-AgI-P_2O_5$
16	Oxynitride	$MgO-Al_2O_3-AlN-SiO_2$

[0243]

<Method of producing the organic EL element>

Next, the method of producing the organic EL element of the present invention is described. The anode is preferably produced by the vapor-growth method, e.g., evaporation or sputtering.

[0244]

The cathode can be produced by evaporation or sputtering, the former being more preferable in consideration of making the film on the organic layer, because it damages the organic layer less.

[0245]

For production of the organic layer, e.g., light-emitting layer, vacuum evaporation is the preferable method for its capacity of forming uniformly thin films. Vacuum evaporation gives a uniformly thin film, amorphous or having a grain size of $0.1\mu\text{m}$ or less, the lower limit normally being around $0.001\mu\text{m}$. The grains having a size exceeding $0.1\mu\text{m}$ may cause problems, e.g., uneven emission of light, high driving voltage required, and greatly reduced charge injecting efficiency.

[0246]

The vacuum evaporation conditions are not limited, but degree of vacuum and deposition rate are preferably 10^{-3}Pa or less and 0.1 to 1nm/second . At the same time, each layer is preferably formed continuously under a vacuum, because this

prevents adsorption of impurities on the interface between the layers to secure good characteristics, and, at the same time, reduces driving voltage of the element and controls generation/growth of dark spots.

[0247]

When vacuum evaporation is used to have a mixed layer or the like which contains 2 or more compounds in one layer, it is preferably formed by coevaporation, in which each compound carried by a boat is individually temperature-controlled to be evaporated by an independent evaporation source. However, when vapor pressures (evaporation temperatures) of the different compounds are similar or very close to each other, they may be mixed beforehand in the evaporation boat and then evaporated.

[0248]

The other methods include solution coating (spin coating, dip coating, casting, etc), and Langmuir-Blodgett (LB) method. The solution coating method may be structured in such a way that each compound is dispersed in the matrix material (resin binder), e.g., polymer. The method of producing the color filter is described earlier.

[0249]

The organic EL element of the present invention is normally DC-driven, but can be AC- or pulse-driven. The voltage to be applied is normally in a range of around 2 to 10V, which is

lower than that for the conventional device.

[0250]

[EXAMPLES]

The present invention is described in more detail by EXAMPLES and REFERENCE EXAMPLES. The compounds used in EXAMPLES are described below by their structural formulae.

[0251]

Tetraarylbenzidine derivative (No. I-1)

Phenyl anthracene derivative (No. I-1)

[0252]

Naphthacene derivative (No. 20)

Styryl amine derivative (S-9)

N,N'-di(1-naphthyl-N,N'-diphenyl benzidine (NPB)

[0253]

<EXAMPLE 1>

A glass substrate was coated with a 100nm thick film of ITO as the transparent electrode (anode) by sputtering.

[0254]

The glass substrate coated with the transparent electrode of ITO was washed ultrasonically with a neutral detergent, acetone and ethanol. It was withdrawn from boiling ethanol, dried, washed with UV/O₃, and set on a substrate holder in a vacuum evaporation apparatus. The vacuum tank was evacuated to 1×10^{-4} Pa or less.

[0255]

Then, N,N'-diphenyl-N,N'-bis[N-phenyl-N-4-tolyl (4-aminophenyl)]benzidine (HIM34) was deposited at 0.2nm/second by evaporation to a thickness of 20nm, to form the hole injecting layer.

[0256]

Next, a N,N,N',N'-tetrakis-(3-biphenyl-1-yl)benzidine (tetraarylbenzidine derivative (No. I-1) was deposited at 0.2nm/second by evaporation to a thickness of 20nm, to form the hole transferring layer.

[0257]

Next, a tetraarylbenzidine derivative (No. I-1), 10,10'-bis[2-biphenyl]-9,9'-bianthryl(phenyl anthracene derivative (No. I-1)) and naphthacene derivative (No. 20) were deposited by coevaporation to a thickness of 30nm, to form the first light-emitting layer of mixed layer type having the volumetric tetraarylbenzidine derivative/10,10'-bis[2-biphenyl]-9,9'-bianthryl(phenyl anthracene) derivative ratio of 1:3 and containing the naphthacene derivative at 3.0% by volume. These compounds were deposited at 0.05, 0.15 and 0.006nm/second, respectively.

[0258]

Next, a tetraarylbenzidine derivative (No. I-1), phenyl anthracene derivative (No. I-1) and styryl amine derivative (S-9) were deposited by coevaporation to a thickness of 50nm, to form the blue-emitting layer as the second light-emitting

layer of mixed layer type having the volumetric tetraarylbenzidine derivative/phenyl anthracene derivative ratio of 1:3 and containing the styryl amine derivative at 3.0% by volume. These compounds were deposited at 0.05, 0.15 and 0.006nm/second, respectively.

[0259]

Next, phenyl anthracene derivative (No. I-1) was deposited at 0.05nm/second by evaporation to a thickness of 20nm, while keeping the system under a vacuum, to form the electron transferring layer. Moreover, tris(8-quinolinolate)aluminum (AlQ3) was deposited at 0.2nm/second by evaporation to a thickness of 10nm, to form the electron injecting layer.

[0260]

Next, CsI and MgAg (10:1 by weight) were deposited in this order at 0.05 and 0.2nm/second by evaporation to a respective thickness of 0.2 and 200nm, while keeping the system under a vacuum, to form the cathode, which was coated with a 100nm thick aluminum as the protective layer by evaporation, to prepare the organic EL element.

[0261]

The resultant organic EL element comprises, as shown in Figure 1, the substrate 1 coated with the anode 2, which supports the hole injecting layer 3, hole transferring layer 4, first emitting layer 5 of mixed layer type, second emitting layer

6 of mixed layer type, electron transferring layer 7, electron injecting layer 8, and cathode layer composed of the lower layer 9 of an alkali metal compound and upper layer 10 of a metal having a low work function, in this order, to emit light from the substrate 1 side.

[0262]

The organic EL element, when operated at a constant current density of $10\text{mA}/\text{cm}^2$, emitted white-color light of $1,100\text{cd}/\text{m}^2$ as the initial luminance at a driving voltage of 6.0V. The half-life of the luminance was 600 hours under the conditions of constant current density: $100\text{mA}/\text{cm}^2$, initial luminance: $9,000\text{cd}/\text{cm}^2$ and driving voltage: 9.9V, and 50,000 hours under the conditions of constant current density: $10\text{mA}/\text{cm}^2$, initial luminance: $1,100\text{cd}/\text{cm}^2$ and driving voltage: 6.0V.

[0263]

<EXAMPLE 2>

The organic EL element was prepared in the same manner as in EXAMPLE 1, except that the tetraarylbenzidine derivative (No. I-1) was replaced by N,N'-di(1-naphthyl)-N,N'-diphenyl benzidine (NPB), and its characteristics were analyzed also in the same manner. It emitted light of white color. The half-life of the luminance was 500 hours under the conditions of constant current density: $100\text{mA}/\text{cm}^2$, initial luminance: $9,000\text{cd}/\text{cm}^2$ and driving voltage: 9.8V, and 35,000 hours under the conditions of constant current density: $10\text{mA}/\text{cm}^2$, initial

luminance: 1,100cd/cm² and driving voltage: 5.5V.

[0264]

<EXAMPLE 3>

The organic EL element was prepared in the same manner as in EXAMPLE 1, except that the First light-emitting layer of mixed layer type was not provided, and its characteristics were analyzed also in the same manner. It emitted light of blue color. The half-life of the luminance was 500 hours under the conditions of constant current density: 100mA/cm², initial luminance: 8,500cd/m² and driving voltage: 7.8V, and 30,000 hours under the conditions of constant current density: 10mA/cm², initial luminance: 900cd/m² and driving voltage: 6.0V.

[0265]

The spectral patterns of the light emitted from the organic EL elements prepared in EXAMPLES 1 and 3 are shown in Figure 2. They emitted light of white and blue color, respectively.

[0266]

<REFERENCE EXAMPLES 1>

The organic EL element was prepared in the same manner as in EXAMPLE 1, except that cesium iodide for the electron injecting electrode was not used and MgAg was directly deposited on AlQ3 by evaporation to form the electrode.

[0267]

It emitted light of orange color having a luminance of 400cd/m² at a constant current density of 10mA/cm² and driving

voltage of 9.0V. The spectral pattern of the emitted light was measured. It was found that 90% or more of the light was from the naphthacene derivative (No. 20).

[0268]

Its light-emitting life was measured. It emitted light having a luminance of 4,000cd/m² at a constant current density of 100mA/cm². Its half-life was 4 hours. It was particularly noted that the blue light greatly lost its luminance.

[0269]

<REFERENCE EXAMPLES 2>

The organic EL element was prepared in the same manner as in EXAMPLE 1, except that the tetraarylbenzidine derivative (No. I-1) and phenyl anthracene derivative (No. I-1) as the host compounds for the mixed layer type light-emitting layer was replaced by phenyl anthracene derivative (No. I-1) as the single host compound, which was doped with the naphthacene derivative (No. 20) and styryl amine derivative (S-9) in a similar manner.

[0270]

It emitted light of orange color having a luminance of 900cd/m² at a constant current density of 10mA/cm², when operated at a driving voltage of 7.5V. The spectral pattern of the emitted light was measured. It was found that 75% or more of the light was from the naphthacene derivative (No. 20).

[0271]

SECRET 2/3/2005

Its light-emitting life was measured. It emitted light having a luminance of $9,000\text{cd/m}^2$ at a constant current density of 100mA/cm^2 . Its half-life was 100 hours. It was particularly noted that the blue light greatly lost its luminance.

[0272]

<EXAMPLE 4>

[Preparation of the organic EL element]

A glass substrate (Corning's 7059) was scrubbed with a neutral detergent.

[0273]

The substrate was coated with pigment-dispersed type color filters, which is the most common procedure to colorize a liquid crystal display, under the coating conditions to have the 1.0 to $1.5\mu\text{m}$ thick film for each of the red, green and blue colors. Each filter was treated to have the desired pattern. The substrate was coated with the red color filter material by spin coating at $1,000\text{rpm}$ for around 5 seconds, and the resultant color filter was prebaked at 100°C for 3 minutes. The photomask was aligned by an exposure device, irradiated with ultraviolet ray of 20mW for around 30 seconds, and developed with an around 0.1% by weight aqueous solution of tetramethyl ammonium hydride (TMAH) for around 1 minute. It was cured at 220°C for 1 hour to form the red color filter. The curing was to prevent the color filter from dissolving the color filter solution of another color, to be spread thereon subsequently.

The color filters were prepared on the red color filter one after another by the similar procedure, although the detailed conditions for forming the filters were different for the different materials (pigments).

[0274]

Next, the color filter was coated with an overcoat of acrylic resin to have the flat surface for the ITO film to be provided thereon. The overcoat material was treated to have a desired pattern, and cured at around 220°C for 1 hour, to have the overcoat layer, around 3μm thick.

[0275]

Next, ITO was spread on the overcoat layer by sputtering to form the transparent, electroconductive layer, around 100nm thick. It was treated by photolithography to have a resist pattern, and etched with a diluted hydrochloric acid. Then, the resist was removed, to have the ITO pattern.

[0276]

The patterned ITO layer was coated with a SiO₂ layer as the insulation film by sputtering. It was patterned in such a way to leave SiO₂ in the area other than the area where the emitted light could be seen from the substrate side, to form the insulation film, around 0.1μm thick.

[0277]

The organic layer for the organic EL element, cathode and protective layer were formed in the same manner as in EXAMPLE

1, to prepare the organic EL displays having the white, green or blue dots. The picture element was 2 by 2mm in size, and number of the picture element was 1 dot for each color.

[0278]

Each of the organic EL element displays, when operated at a constant current density of $100\text{mA}/\text{cm}^2$, was confirmed to emit the color of the following luminance and CIE chromaticity:

[0279]

	Luminance cd/m^2	CIE x/y
White	5260	0.32/0.34
Red	934	0.62/0.34
Green	2900	0.31/0.50
Blue	744	0.12/0.14

[0280]

<EXAMPLE 5>

[Preparation of the simple matrix type organic EL color display]

The substrate prepared in the same manner as in EXAMPLE 4 was set on the substrate holder in a sputtering unit, and coated with an approximately $1.5\mu\text{m}$ thick Al film and then with approximately 30nm thick TiN continuously, to form the laminate of Al and TiN. The Al and TiN films were brought into close contact with each other with no natural oxide film in-between, because they were continuously formed without breaking the vacuum. The laminate film was treated by photolithography to have the low-resistance wiring pattern.

[0281]

The color filter and overcoat layers were formed in the same manner as in EXAMPLE 4. The overcoat layer was patterned in such a way to expose the TiN layer surface.

[0282]

Then, the ITO layer as the transparent electrode was formed in the same manner as in EXAMPLE 4. This connected the low-resistance Al wiring pattern on the ITO layer to that formed beforehand, to form the column lines.

[0283]

The patterned ITO layer was coated with a SiO_2 layer as the insulation film by sputtering. It was patterned in such a way to leave SiO_2 in the area other than the area where the emitted light could be seen from the substrate side, to form the insulation film, around $0.1\mu\text{m}$ thick. The insulation layer worked to avoid useless light emission in an area unseen from the substrate surface. It prevents another trouble; the area has holes or grooves, and the organic EL layer deposited on the inclined portion would become thinner tending to cause current leakage without the insulation layer.

[0284]

The insulation layer was then coated with a solution adjusted to contain 15% by weight of polyimide to a film thickness of $2\mu\text{m}$ by spin coating. The resultant film was prebaked at 145°C for 1 hour, to form the intermediate spacer film.

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Subsequently, it was coated with a positive resist layer, which was treated by exposure and development to have a desired photopattern, in order to form the shade-like photosensitive resin shape. The developing solution removed the positive resist. It also treated the intermediate spacer film into the final shape. This resulted in the divided structure of the element.

[0285]

Next, the organic layer for the organic EL element, cathode and protective layer were formed in the same manner as in EXAMPLE 1, to prepare the simple matrix type organic EL display. Its picture element was 330 by 110 μ m in size, and number of the picture element was 320 by 240 by RGB dot.

[0286]

When operated in line sequence, it emitted colored light having CIE chromaticity levels similar to those observed in EXAMPLE 4.

[0287]

<EXAMPLE 6>

The display was prepared in the same manner as in EXAMPLE 4, except that a black matrix was provided between the aligned color filters. When operated in a similar manner, it emitted sharper light in comparison with EXAMPLE 4. The black matrix was of a common pigment-dispersed type.

[0288]

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<EXAMPLE 7>

The display was prepared in the same manner as in EXAMPLE 4, except that an overcoat layer of acrylic resin and approximately 60nm thick SiO₂ layer as the protective film were provided in this order. When operated in a similar manner, it produced the results similar to those observed in EXAMPLE 4. It was also found that the element had improved durability.

[0289]

[ADVANTAGE OF THE INVENTION]

The present invention efficiently emits light of blue color. It is applicable to the multi-color emission, including blue color, and gives the organic EL element of high luminance and long service life. Moreover, the organic EL element is applicable, with its excellent characteristics, to the organic display emitting multi-colors, when combined with a color filter.

[BRIEF DESCRIPTION OF THE DRAWINGS]

[Figure 1] outlines the cross-section of the organic EL element prepared in EXAMPLES.

[Figure 2] shows the spectral patterns of the light emitted from the organic EL element prepared in EXAMPLES.

[DESCRIPTION OF THE REFERENCE NUMERALS AND SIGNS]

- 1 Substrate
- 2 Anode
- 3 Hole injecting layer

- 4 Hole transferring layer
- 5 First emitting layer
- 6 Second emitting layer
- 7 Electron transferring layer
- 8 Electron injecting layer
- 9 Lower layer of cathode
- 10 Upper layer of cathode

[Figure 2]

Strength (arbitrary unit)

Wavelength (nm)